ISSN 0840-8440

PROCEEDINGS

TECHNOLOGY TRANSFER CONFERENCE 1988

November 28 and 29, 1988

Royal York Hotel

Toronto, Ontario

SESSION A
AIR QUALITY RESEARCH

Sponsored by

Research and Technology Branch

Environment Ontario

Ontario, Canada

AA SG

A7

HAZARDOUS CONTAMINANTS IN ONTARIO:

ENVIRONMENTAL FATE AND

HUMAN EXPOSURE

by

D. Mackay

S. Paterson

W. Y. Shiu

Institute for Environmental Studies

University of Toronto

Toronto, Ontario, M5S 1A4, Canada

A paper to be presented at the 1988 Annual Technology

Transfer Conference, November 28-29, 1988

ABSTRACT

During recent years our group has been developing and testing various models which describe the environmental fate of hazardous chemicals in Southern Ontario. The current status of these models is reviewed. A capability now exists of estimating the prevailing concentrations of such chemicals in air, water, soil and sediments provided that physical chemical properties and emission rates are available. The models can vary in region of application, in complexity and can be steady and unsteady state in nature. In recent work, these models are being extended to assess the magnitude of exposure of a typical Ontario family by routes of air inhalation and water and food ingestion. The principal challenge in this work is quantifying the air-soil-vegetation-meat and dairy products route. Finally, progress is being made in developing pharmacokinetic models which enable these "external" exposures to be translated into concentrations or "internal" exposures in various human tissues. It is thus becoming possible to quantify the entire journey of a toxic chemical from emission source to human target organ and explore how these target concentrations will respond to various strategies of emission or loading reduction.

INTRODUCTION

During the last decade various environmental models have been developed to describe or predict the multimedia fate and behavior of chemicals (1-5). These models vary in complexity and may be steady- or unsteady-state in nature. The models accept data on the chemical's physical chemical properties, reactivity, transport characteristics, and extent of release to the environment to produce a comprehensive picture of the chemical's pathways between, and ultimate concentrations in, various media such as

soil, air, water, and sediment. This modelling capability is a necessary adjunct to toxicity testing when assessing risk from chemical exposure and for determining priorities for research and regulatory action. It can be applied to new and existing chemicals. A major purpose of such models is to produce lists of priority chemicals such as Ontario's "EMPPL" list.

Of most concern are chemicals that are, or may be, in frequent commercial use and therefore will be discharged constantly to the environment over a period of time. Examples are pesticides, PCB's, wood preservatives, and by-products of incineration.

The use of the fugacity approach in some of these models (1,2) simplifies the algebra and clarifies the interpretation. This approach has been used by other groups for example, in the development of the OECD Environmental Exposure Potentials (EEP) model, ECETOC assessments, the U.S. EPA ENPART model (which uses the model as a first level screening method to predict the environmental fate and human exposure potential of new and existing chemicals), the National Institute of Public Health and Environmental Hygiene (RIVM) in the Netherlands and the Mitsubishi-Kasei Institute of Toxicological and Environmental Science in Yokohama (6-12).

In this paper we describe recent studies in our group supported by the Ontario Ministry of Environment which have sought to develop, validate and implement environmental models of the multimedia fate of chemicals. Further, we describe current efforts, hopes for the future, and speculate on how environmental models may evolve as regulatory and scientific tools. First, we describe the development of a novel fugacity model.

THE FOUR MEDIA FUGACITY MODEL

Previous models have described the fate of chemicals in a multitude of media such as air, aerosols, water, fish, particles, sediment, sediment pore water, etc. From an examination of this situation, it has become clear that we can simplify the model with little loss of fidelity by treating only four bulk phases - air, water, soil and sediment, which consist of subcompartments of fluid (air and/or water), solid and biotic matter. Equilibrium partitioning of chemicals is assumed to apply within each of the four bulk phases. As a result only four fugacities are defined, but it is possible to calculate concentrations in subcompartment phases.

Expressions for environmental emissions, advective inflow and outflow, reaction, and diffusive and non-diffusive transfer between these bulk phases are incorporated in a set of mass balance equations as included in previous models. Reducing the number of key environmental media to four permits a simple algebraic solution which can be implemented on a personal computer. Unlike previous models, non-diffusive transfer processes, such as wet and dry atmospheric particulate deposition, rain washout, leaching to groundwater from soil, and sediment resuspension are included.

<u>Fugacity</u>
In this model partitioning of chemical between phases is described by the equilibrium criterion of fugacity as an alternative to chemical potential.

Fugacity can be considered to be the partial pressure of a chemical in a phase and is related to its concentration in that phase by the expression

$$C = fZ$$

where C is concentration (mol/m^3), f is fugacity (Pa) and Z is the fugacity capacity (mol/m^3.Pa) which is specific to the chemical, the medium and temperature. Z values are deduced from physical chemical properties such as molecular mass, solubility, vapour pressure and octanol-water partition coefficient. Methods of calculating Z values for environmental phases have been reviewed by Mackay and Paterson (1). This data set represents an absolute minimum for chemical fate assessment.

Table 1 lists the properties of six priority chemicals which have been the subject of assessment in this project. A significant part of our efforts has been devoted to measuring and correlating these properties which are of fundamental importance in any fate assessment.

Chemicals move between phases by diffusive and non-diffusive processes. The diffusive flux N (mol/h) between two phases, 1 and 2, can be described by

Phase 1 to 2 rate = $D_{12}f_1$

Phase 2 to 1 rate = $D_{21}f_2$

Net rate N = $D_{12}f_1 - D_{21}f2$

where D_{12} and D_{21} are transfer coefficients with units of mol/h.Pa, and f_1 and f_2 are the phase fugacities. The difference between f_1 and f_2

determines the direction of diffusive flux (but not non-diffusive flux) which takes place from high to low fugacity. The coefficients D_{12} and D_{21} are functions of Z values, interfacial areas and diffusion properties, (characterized by diffusivities or mass transfer coefficients) in adjacent phases (2).

Non-diffusive or one-way transfer processes between phases, for example, wet or dry particle deposition from the atmosphere to soil or water, or suspended sediment deposition or resuspension, is described by a one-way transport parameter as

$$N = GC = GZf = Df \text{ mol/h}$$

where G is the volumetric flow rate (m^3/h) of the transported material (eg sediment), and D again has units of mol/Pa.h.

First order reaction processes in a phase can be described by

$$\text{rate of reaction} = k_R VC = k_R VZf = D_R f \quad \text{mol/h}$$

where k_R is a first order rate constant (h^{-1}), V is the phase volume (m^3), and D_R again has units of mol/Pa.h.

In summary all rates of chemical transport and transformation can be expressed as products of fugacities and D values. D values can be summed and compared when the processes apply to the same source phase fugacity.

Environmental Media

Four major bulk compartments are defined - air, water, soil, and sediment consisting of a combination of subcompartments of pure and particulate phases of defined volume fractions. For example, bulk water consists of "pure" water, suspended solids, and fish; soil consists of mineral matter, organic matter, water, and air; air consists of pure air and aerosols; sediment is mineral matter, organic matter, and water.

This environment can be "evaluative" or purely hypothetical in nature as described in previous papers (3,13) or it can represent an actual region. We apply the model here to i) an environment scaled to represent Southern Ontario and ii) an evaluative environment of area 1 sq km with similar phase proportions. The air height has been reduced to 2000 m from 6000 m to reflect conditions of atmospheric accessibility in Southern Ontario.

The assumption that equilibrium exists within each bulk phase, (ie. a common fugacity applies) implies that the times required to reach equilibrium <u>within</u> a bulk phase are short compared to times required to reach equilibrium <u>between</u> bulk phases.

Levels of Model Complexity

Environmental scientists compete within the arena of the refereed scientific literature to produce models which are more reliable, comprehensive and valid. A consequence of this pressure is a tendency to

produce increasingly complex models - an understandable direction because the environment is a very complex system.

This produces a problem because individuals in industry and government who use these models have relatively little time to learn the intricacies of the models and acquire the necessary input data. Further, the more conscientious are reluctant to use and trust models which they do not fully understand. We believe that the answer is to offer a series of models, varying in scope, complexity and data needs. Presumably as users become more familiar with the simple models they will become increasingly dissatisfied with their simplifying assumptions and move towards more complex models. Situations of greatest concern which merit most effort will justify the most detailed assessment using the most complex models. Often a simple model will give an adequate answer to a simple question. Accordingly we have developed a series of models, increasing in complexity.

Level I Calculations

Using the phase volumes and Z values (obtained from physical-chemical properties) and an assumed total amount of chemical present, M mols, it is possible to calculate its equilibrium partitioning in this multimedia environmenta. A mass balance gives

$$M = \sum V_i C_i = f \sum V_i Z_i$$

The prevailing fugacity can thus be calculated as $M/\sum V_i Z_i$, followed by bulk concentrations, individual subcompartment concentrations (as Zf) and amounts as CV or VZf. This provides a picture of the dominant media into which the chemical partitions and provides a first approximate estimate of relative concentrations. Table 2 gives an example of such calculations.

The primary difficulty is estimating the amount of chemical M. Two approaches can be used. First is to gather concentration data for various media and calculate and sum amounts for the region of interest. Second, if an emission or use rate is available eg. 1000 kg/year and an approximate environmental persistence can be estimated, eg. 0.3 years, then the amount present will be the product of these quantities or 300 kg. If concentration data are available for one or two media (eg. fish and air), it is possible to select M by trial and error to obtain the reported values for fish and/or air. There is usually order-of-magnitude uncertainty about M.

This simple model does not treat reactions, inflow in air and water and other loss processes.

TABLE 1: PHYSICAL CHEMICAL PROPERTIES OF SELECTED PRIORITY CHEMICALS AT 25°C.

Chemical	Mol. Mass g/mol	Vapor Pressure Pa	Solubility g/m^3	$\log K_{OW}$
Hexachlorobiphenyl*	350	5×10^{-4}	3.5×10^{-3}	6.8
benzene	78	12700	1780	2.13
benzo a pyrene	252	7.3×10^{-7}	3.8×10^{-3}	5.98
hexachlorobenzene	285	1.5×10^{-3}	5.0×10^{-3}	5.47
mirex	322	2×10^{-4}	1.93×10^{-5}	6.8
trichloroethylene	131.5	9870	1100	2.29

* average properties of isomers

TABLE 2: LEVEL I CALCULATION OF PCB DISTRIBUTION IN SOUTHERN ONTARIO

Total amount 3×10^{6} mol or 1.07×10^{6} kg

Fugacity 1.8×10^{-7} Pa

Compartment	Volume m^3	Conc.	Amount mol.	%
Bulk Air	4×10^{14}	3 ng/m^3	36000	1.2
Bulk Water	4×10^{12}	9.5 ng/L	109000	3.5
Bulk Soil	1.2×10^{10}	50 ng/g	2.7×10^{6}	88
Bulk Sediment	8×10^{8}	60 ng/g	216000	7.0

Subcompartment example:

Air-gaseous	2.5 ng/m^3	0.95 %
Air-aerosol	0.5 ng/m^3	0.23 %

Level II Calculations

In this model two loss processes are considered, advection ie. loss by outflow mostly in air or water, and degrading reactions. Also included are three "pseudo advection" processes. Transfer from air to higher altitude can be assumed to occur at a specific velocity. Leaching from soil to groundwater can be set at a percentage of the rain rate on soil. Sediment burial rates can be assumed and will equal the difference between sediment desposition and resuspension rates. Values suitable for specific regions can be inserted.

Reaction rates are included as first order rate constants applicable to the bulk or pure phase. These rate constants or half-lives are chemical-specific and usually result from a consideration of the rates of processes such as biodegradation, photolysis, oxidation and hydrolysis, individually and in total.

If emission rate data are available, a steady state, equilibrium calculation can be completed. The emissions rate should include point and nonpoint sources, and to it should be added advective inflow at a suitable "background" concentration or fugacity. From a mass balance, the prevailing fugacity can be calculated, followed by concentrations, amounts and inflow and loss rates. It is illuminating to examine the relative importance of these loss processes. The total amount present M can be calculated and used to estimate the chemical's overall "persistence" or average residence time. This time is a combination of the reaction time or persistence and the advection residence time.

Table 3 gives an illustrative calculation for PCBs in Southern Ontario. This behavior profile contains the assumption that a common fugacity prevails throughout the environment. Clearly this is in error because fugacities tend to be higher in the media which receive direct discharges and lower in those which have relatively rapid loss mechanisms. In particular, it appears that the air often experiences a low fugacity because of the high rate of advective loss. To remedy this assumption it is necessary to allow for fugacity variation between media and include expressions for interphase transport conductivities or resistances and introduce the emissions on a medium-specific basis as in the more complex Level III calculation.

Level III Calculations

Steady state mass balance equations can be set up for each of the four bulk phases which incorporate emissions, transfer rates between adjacent phases, reaction and advection. Four algebraic equations result which can be solved simultaneously. The principal difficulty is to estimate the inter-media transfer rate parameters or D values. These D values depend on the Z values (which are chemical specific) and on other kinetic quantities such as mass transfer coefficients or deposition rates of suspended matter.

It is important to emphasize that in the model single typical values have been assumed for all kinetic terms, ie. they are not chemical specific. There is thus no need to include more chemical-specific data in the Level III calculation. The dependence of transfer rate on chemical properties is

TABLE 3: LEVEL II CALCULATION OF PCB IN SOUTHERN ONTARIO

Fugacity	2.8×10^{-8} Pa		
Input emissions	45 mol/h	Advection -	1 mol/h
Residence time	1.2 years		
Amount	4.8×10^{5} mol		

		Loss Rates (mol/h)	
Compartment	Conc.	Reaction	Advection
Bulk Air	0.5 ng/m^3	-	47
Bulk Water	1.5 ng/L	0.03	1.4
Bulk Soil	8 ng/g	6.4	10^{-4} (leaching)
Bulk Sediment	10 ng/g	0.5	0.4 (burial)

TABLE 4: SELECTED REPORTED AND CALCULATED CONCENTRATIONS FOR TEST CHEMICALS USING LEVEL III MODEL. Calculated values in parentheses.

Chemical	
PCB	Air 1-10 (1.4) ng/m^3: Water 1-10 (5) ng/L
	Fish 0-1.7 (0.2) ug/g
Benzene	Air 16 (2.5) ug/m^3
BaP	Air 1.3-7.1 (3.0) ng/m^3: Soil 0.1-0.5 (0.04) ug/g
	Sediment 80-300 (190) ng/g
HCB	Air 0.2 (0.2)* ng/m^3: Water 0.05-1 (2.7) ng/L
	Fish 0.003-0.05 (0.1) ug/g
TCE	Air 6.4 (11) ug/m^3
Mirex	Water 0.002-30 (0.002) ng/L
	Fish 0-0.04 (0.0001) ug/g

* Emissions adjusted to give reported air concentrations

contained entirely in the Z values which comprise part of the D values. No claim is made that the assumed coefficients are applicable to other environments, indeed there is considerable doubt about some of the values in Southern Ontario. The user is encouraged to modify these values in the light of experience. The values given should be regarded as only reasonable first estimates.

This model contains approximately 40 environment-specific parameters (volumes, velocities, diffusivities etc.) which are assumed to apply to all chemicals. In many cases the values are not well established (eg sediment deposition rate), thus the user should regard these parameters as initial order-of-magnitude estimates which will be subject to change in the light of experience. Reaction rate constants are both chemical and environment specific. Finally emission rates are region-specific and are often the most poorly quantified of all variables.

Reasonable parameter values have been assembled for the Southern Ontario - lower Great Lakes Basin Region. They can be modified for other regions. The differences between the regional and evaluative models are the volumes, areas and advective flow rates. All other rates are expressed on a unit area or volume basis and thus apply to both models.

The level III output can include all fugacities, concentrations (in mol/m^3 and conventional units) and transfer rates for individual and combined processes. A total mass balance can be assembled, as illustrated for PCBs in Figure 1, and the overall chemical persistence can be calculated.

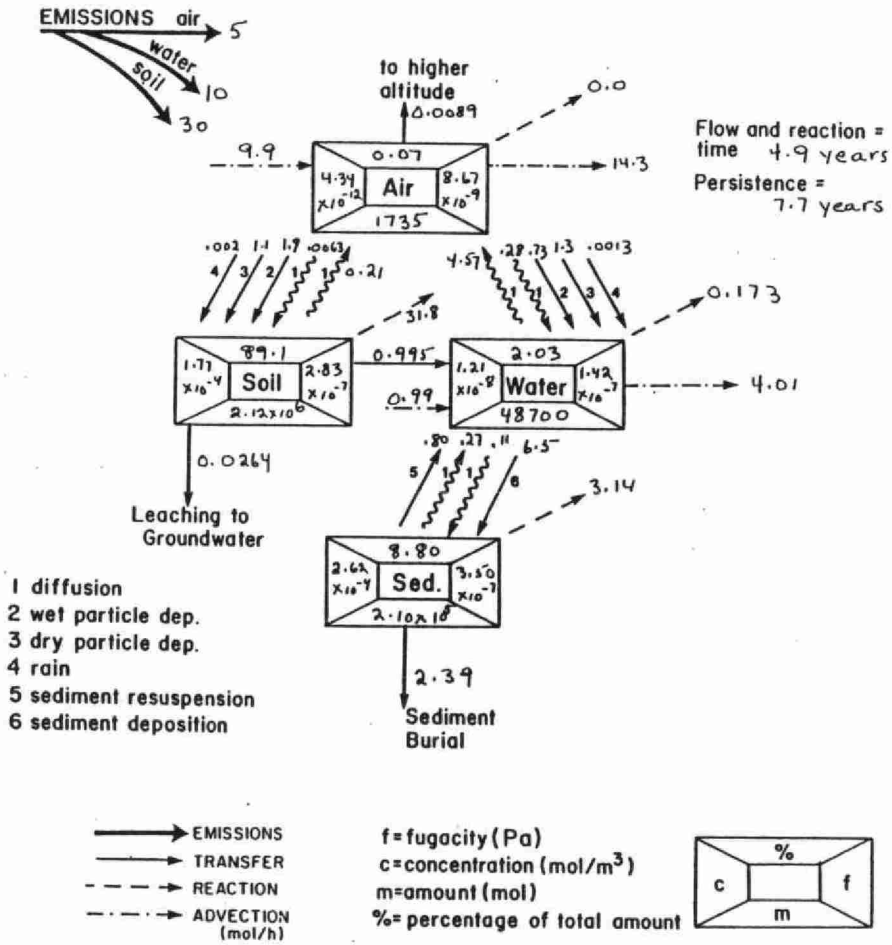

FIGURE 1: Level III mass balance for PCBs in Southern Ontario

"VALIDATION" USING TEST CHEMICALS

Models such as this cannot be "validated" in the same sense that a simpler physical or chemical model can be validated. Environmental concentrations vary in time and space, and few reliable data are usually available. Reaction rate constants vary diurnally and seasonally. Emissions are rarely known accurately. There are variations and uncertainties in transport rate parameters, such as depositiion and resuspension rates. Few environments are at steady state. The best that can be hoped for is that the model corresponds with order of magnitude fidelity to fragmented observations for a variety of chemicals of quite different properties and pathways. The chemical-to-chemical variation should be described entirely by the chemicals' properties and emission rates and not by chemical-specific adjustable parameters. But even this modest model capability is invaluable for predicting the behavior of new chemicals and for estimating order of magnitude concentrations.

The model was run for selected chemicals (Table 1) of varying properties, and environmental concentrations were calculated, and compared with observations. A common problem is estimation of emission rates. In some cases, it is necessary to back-calculate what the emissions may be, or must be, to create prevailing concentrations, then check if these emission rates are in reasonable accord with rates of production or use.

Table 4 lists some illustrative calculated bulk compartment concentrations and fugacities and gives reported values for comparison. There is generally order-of-magnitude agreement.

DISCUSSION

The set of models provides a relatively simple, rapid method of establishing the relative importance of environmental fate processes for specific chemicals using minimal data. It is believed to give an adequate characterization of the dominant phases of accumulation. For example, mirex is 62% in soil and 35% in sediments, while benzene is 46% in air and 54% in water. These percentages are controlled by the nature of the emission sources and the chemicals' properties. An overall persistence or residence time can also be estimated which ranges from 6 days for benzene to 12 years for mirex. This persistence is a function of partitioning, and rates of reaction and advection. The relative importance of advective inflow and local emissions can be estimated.

The evaluative version can be used to determine the dominant environmental partitioning pathways and the persistence of a chemical, using illustrative rather than real emissions. The computed absolute concentrations then have no significance. Since many jurisdictions have a common interest in estimating the environmental fate of chemicals, but the regions corresponding to these jurisdictions have differing ratios and natures of air, water, soil, and sediments, it is useful to have a common evaluative test system which can be used to inter-calibrate environmental assessments. The combination of evaluative and region-specific environments thus provides a convenient system for international assessment of chemicals.

A useful feature of both models is that a sensitivity analysis is possible by which the effects of varying properties of the chemicals and the

environment can be explored. This establishes which properties should be estimated most accurately and can provide an estimate of likely error in concentration as a function of error in property.

If emission estimates are available, the model can be used to estimate concentrations with, it is believed, order of magnitude (factor of 10) accuracy. It is most reliable for substances, such as PCBs, which are persistent and widely dispersed in the environment. The model can be used as an interpretive tool to explore temporal and spatial variations in concentrations. For example, it appears that mirex has been discharged over two time periods at emission rates which have differed by a factor of approximately 500. Since the model is linear, concentrations are linearly related to total emissions, thus there is no need to run the model repeatedly to explore such changes. Because certain contaminants, such as benzene and BAP are emitted with considerable spatial variation, it can be misleading to use average emissions and concentrations. It may be possible to develop "rules", which may be region-specific, that one would expect to encounter local concentrations which are perhaps a factor of 10 or 20 higher than average.

The model has been described in steady state form, but with little additional effort it can be written in differential equation form and solved numerically to give the response in the region to time-varying emissions. This is a Level IV calculation.

These estimated concentrations are of potential value in three respects.

First, they provide guidance about likely prevailing environmental concentrations in the various media which result from present or possible future emissions. This is useful when designing analytical monitoring and identification programs. Second, they provide information about the proximity of the concentrations to those which are judged to be of toxicological or aesthetic significance.

Third, the concentrations provide a starting point for estimation of human exposure. These issues are discussed more fully in the next section.

FROM CONCENTRATIONS TO EXPOSURE AND EFFECTS

Exposure assessment calculations can take one or both of two directions; comparison with targets, and dosage assessment.

"Target" concentrations may be defined for each medium. For example, from considerations of toxicity or aesthetics it may be possible to suggest that water concentrations of a specific chemical should be maintained below 1 ug/litre, air below 1 ug/m^3, and fish below 1 mg/kg. These target concentrations can then be compared as a ratio or quotient to the estimated environmental concentrations. A hypothetical example is given in Table 5 showing the application of this "quotient" method. In this case it is apparent that the primary concern is with air and fish. The proximities of the estimated prevailing concentrations to the targets are quantified by these quotients, a large value implying a large safety factor. The high-risk situations correspond to low quotients. The concentration level in

fish is not directly toxic to the fish but poses a possible threat to humans if consumed on a regular basis.

There are often problems associated with suggesting target concentrations in soil and sediment because these media are not normally consumed directly by organisms. Whereas simple lethality experiments can be designed using air, water, or food as vehicles for toxicant administration, it is not always clear how concentrations in the solid matrices of soils and sediments relate to exposure or intake of a chemical by organisms. It is difficult to design meaningful bioassays involving interactions between organisms, soils, and sediments.

The second approach is to use these media concentrations to calculate dosages to human and other organisms, as illustrated in Table 5 and Fig. 2 which are reproduced from reference 5.

An average human inhales some 20 m^3 of air per day, thus the associated amount of chemical can be easily calculated. Not all this chemical may be absorbed, but at least a maximum dosage can be evaluated. The same human may consume 2 litres/day of water with its contained chemical, enabling this dosage to be estimated. Food, the other vehicle, is more difficult to treat. An average diet may consist of 1 kg/day broken down as shown in Table 5. Fish concentrations can be estimated directly from the model, but meat, vegetable, and dairy product concentrations are still poorly understood functions of the concentrations of chemical in air, water, soil, animal feeds, and of agrochemical usage. It is likely that techniques will emerge for calculating food - environment concentration ratios, but at

TABLE 5: COMPARISON OF ENVIRONMENTAL CONCENTRATIONS AND HUMAN INTAKE RATES WITH EFFECT LEVELS

Comparison of environmental effect concentrations with predicted concentrations

	Effect level	Actual level	Quotient or ratio
Air pollution ($\mu g/m^3$)	15	2.82	5.3
Fish toxicity (g/m^3)	5	2.91×10^{-4}	17,160
Soil contamination ($\mu g/g$)	5	3.58×10^{-2}	140

Comparison of allowable human intake with predicted intakes[a]

	Concentration (g/m^3)	Volume of intake (m^3)	Actual intake (μg/day)	Percent of permissible intake	Percent of actual intake
Ingestion of food and water					
Fish	0 .0	0.00025	35	0.7	34
Meat products	0.140	0.00005	7	0.14	6.8
Dairy products	2.9×10^{-4}	0.00025	0.073	0.001	0.07
Vegetable products	2.9×10^{-4}	0.00045	0.13	0.003	0.13
Water	2.9×10^{-4}	0.002	0.58	0.012	0.56
Air inhalation	2.82×10^{-6}	20	56	1.1	54.4
Domestic exposure			1	0.02	1
Occupational exposure			3	0.06	3
Total			103[b]	2.04	100

[a]Average daily intake (ADI) = 5000 μg/day.
[b]Ratio of ADI intake = 5000/103 = 48.5.

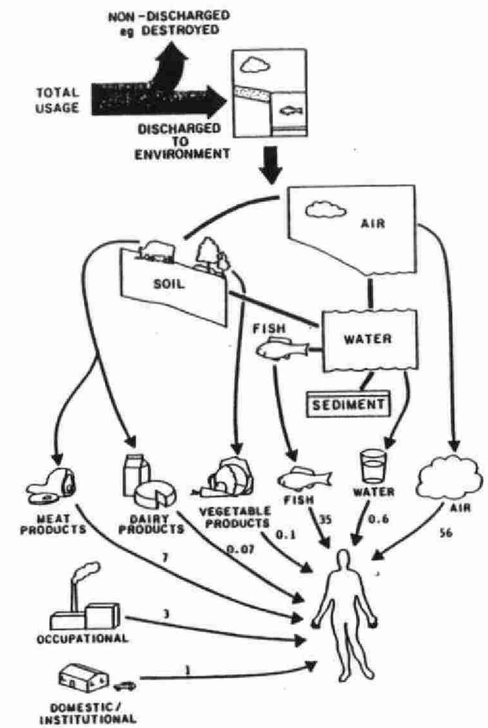

FIGURE 2: Predicted routes of human exposure to a hypothetical compound (ug/day).

present the best approach is to analyze a typical "food basket". The food concentrations used here are illustrative and were obtained by assuming that the meat consumed had partitioning properties similar to fish, and that dairy and vegetable products consisted largely of water. This issue is complicated by the fact that much food is grown at distant locations and imported. Beverages, food, and water may also be treated (deliberately or accidentally) for chemical removal commercially or domestically during food preparation.

It is becoming clear that food ingestion is often the dominant route by which humans are exposed to chemicals, especially agricultural chemicals and those which are emitted into the atmosphere and are subject to wet and dry deposition and absorption on leaf surfaces. This area is the subject of our current efforts in which we are attempting to develop correlations between environmental concentrations (especially air and soil) and those in vegetation, meat and dairy products. Further, we are assembling various models of human intake corresponding to defined population groups with specified diets. One group is young children who may ingest quantities of soil. We hope that it will be possible to identify the more susceptible, more highly exposed groups in a systematic manner and validate these findings by actual market basket and diet measurements.

It is worth emphasising at this point that we do not expect to find that human intake levels will be close to lethal levels, or even to levels of possible concern. But we believe that it is essential for a responsible government to know the principal routes by which the population is exposed to individual priority chemicals. It should also ascertain the proximity

of intake rates to "allowable" levels. When such information is lacking there may be distortion of regulatory actions and scientific efforts into areas which are of high profile in the news media, but in reality are of little concern relative to other more "mundane" issues.

Much chemical exposure may also occur in occupational (e.g., factory) and institutional or commercial (e.g., schools, stores, cinemas) settings, and at home, but these exposures vary greatly from individual to individual and depend on lifestyle. For illustrative purposes and to give a complete picture, exposures from these sources are included in Table 5.

The picture that emerges from this model is a profile of relative exposures by various routes from which the dominant route(s) can be elucidated. If desired, appropriate measures can be taken to reduce certain critical exposures. For example, fish consumption could be reduced. An advantage of this approach is that it places the entire spectrum of exposure routes in perspective. There is little merit in reducing an already small exposure.

CONCLUSIONS

In this project a series of regional fugacity models has been developed, tested with selected priority chemicals, and found to give a reasonable picture of environmental partitioning, reaction, advection, and transport characteristics. Estimated concentrations are in order-of-magnitude accord with reported values, but there are considerable uncertainties in the emission rates on which these concentrations depend. The estimated concentrations may be of value for designing analysis programs, for setting

priorities, and for asssessing human exposure to present and new chemicals.

Our current work is focussing on further validation, developing air-soil-vegetation-meat-dairy product contamination relationships, and in assembling models for human exposure applicable to various groups in Ontario.

We believe that the model can be, and should be, improved by obtaining better estimates of certain transport and partitioning parameters. For example, the mass transfer coefficients and diffusivities are average values applied to all chemicals. Chemical specific and region specific values could be used instead to increase accuracy. Most important, the model should be tested more thoroughly against a variety of chemicals of widely differing properties. The principal difficulties in accomplishing this are the lack of emission data, environmental concentration data, reaction rate constants and physical chemical properties.

The exposure assessment process can be, and should be, extended further to include a physiologically based pharmacokinetic model (15) to predict partitioning, accumulation, and persistence in various animal or human tissues after prolonged, continuous exposure.

Ultimately, a capability is likely to emerge to combine environmental, human exposure, and pharmacokinetic models in an overall process of tracking the pathways of a chemical from its sources, to its distribution in various environmental media, such as air, water, soil, sediment, and food, to its availability or exposure to animals and humans, and finally to target tissues. The model presented here contributes one small step to this process.

REFERENCES

1. Mackay, D., Paterson, S., Environ. Sci. Technol., 1981, 15, 1006-1014.

2. Mackay, D., Paterson, S., Environ. Sci. Technol., 1982, 16, 654-660.

3. Mackay, D., Paterson, S., Cheung, B., Neeley, E.B., Chemosphere, 1985, 14, 335-374.

4. Mackay, D., Paterson, S., in "Pollutants in a Multimedia Environment", Cohen, Y., ed. Plenum: New York, 1986, 117-131.

5. Mackay, D. and Paterson, S., Partitioning Models, pp. 77-86 in Carcinogen Risk Assessment, C.C. Travis (ed)., Plenum Publ. Corp. 1988.

6. ECETOC 1988 Technical Report of the European Chemical Industry Ecology and Toxicology Centre, No. 29 Concentrations of Industrial Organic Chemicals measured in the Environment: The Influence of Physico-Chemical Properties, Tonnage and Use Pattern. Brussels.

7. Klein, A.W., Environmental Exposure Potential (EEP) in Compendium of Environmental Exposure Assessment Methods for Chemicals. OECD Hazard Assessment Panel, Paris, 1988.

8. Wood, W.P., Nold, I.A., Lefler, J., Environmental Partitioning Model (Draft Documentation), Office of Toxic Substances, U.S. EPA, 1982.

9. Calamari, D., Vighi, M., Bacci, E., Chemosphere, 1987, 16, 2359-2364.

10. Zitko, V., McLeese, D.W., Evaluation of Hazards of Pesticides used in Forest Spraying to the Aquatic Environment. Can. Tech. Rept. Fish Aquat. Sci. No. 985, 21pp. 1980.

11. Sloof, W., Cleven, R.G.M.J., Janus, J.A. and Ros, J.P.M. (eds) 1987. Draft Basis Document Copper. RIVM Report No. 758474003, pp. 78-84, RIVM, The Netherlands.

12. Yoshido, K., et al., Toxicol. Environ. Chem., 1987, 15, 159-183.

13. Neely, W.B., Mackay, D., in "Modelling the Fate of Chemicals in the Aquatic Environment", Dickson, K.L., Maki, A.W., Cairns, J. Jr., eds. Ann Arbor Science: Ann Arbor, 1982, 127-143.

14. Mackay, D., Paterson, S., Schroeder, W.H., Environ. Sci. Technol., 1986, 20, 810-816.

15. Paterson, S. and Mackay, D., Environ. Toxic. Chem., 1987, 6, 395-408.